

Form PTO-1300
(Rev. 12-29-99)

US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

ATTORNEY'S DOCKET NO.

H 3381 PCT/US

U.S. APPLICATION NO. (if known) 37 CFR 1.51

097787248

TRANSMITTAL LETTER TO THE UNITED STATES
DESIGNATED/ELECTED OFFICE (DO/EO/US)
CONCERNING A FILING UNDER 35 U.S.C. 371INTERNATIONAL APPLICATION NO.
PCT/EP99/06799INTERNATIONAL FILING DATE
September 14, 1999PRIORITY DATE CLAIMED
September 21, 1998

TITLE OF INVENTION

A SPRAYABLE HOTMELT ADHESIVE

APPLICANT(S) FOR DO/EO/US

Dirk Kolowrot, Heinrich Traeger

Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:

1. ☒ This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.
 2. ☐ This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.
 3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).
 4. ☒ A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
 5. ☒ A copy of the International Application as filed (35 U.S.C. 371(c)(2)).
 - a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☒ has been transmitted by the International Bureau.
 - c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US).
 6. ☒ A translation of the International Application into English (35 U.S.C. 371(c)(2)).
 7. ☒ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
 - a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
 - b. ☐ have been transmitted by the International Bureau.
 - c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
 - d. ☒ have not been made and will not be made.
 8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
 9. ☒ An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). UNEXECUTED
 10. ☐ A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).
- Items 11. to 16. below concern other document(s) or information included:
11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
 12. ☐ An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
 13. ☒ A FIRST preliminary amendment
 - ☐ A SECOND or SUBSEQUENT preliminary amendment.
 14. ☐ A substitute specification.
 15. ☐ A change of power of attorney and/or address letter.
 16. ☐ Other items or information:

"Express Mail" mailing label number EL615774771US

U.S. Application No. (If known, see 37 CFR 1.5) <div style="font-size: 24pt; font-weight: bold; margin-top: 5px;">097787248</div>	INTERNATIONAL APPLICATION NO. PCT/EP99/06799	ATTORNEY'S DOCKET NUMBER H 3381 PCT/IUS			
<p>17. ■ The following fees are submitted:</p> <p>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</p> <p>Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO.....\$1000.00</p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO.....\$860.00</p> <p>International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO\$710.00</p> <p>International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4)\$690.00</p> <p>International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4).....\$100.00</p>		CALCULATIONS PTO USE ONLY			
ENTER APPROPRIATE BASIC FEE AMOUNT = \$ 860					
Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date 37 (CFR 1.492(e)).		\$ 0			
CLAIMS	NUMBER FILED	NUMBER EXTRA	RATE		
Total Claims	1 - 20 =	0	0 X \$18.00	\$ 0	
Independent Claims	1 - 3 =	0	1 X \$80.00	\$ 0	
Multiple dependent claims (s)(if applicable)			0	+ \$270.00	\$ 0
TOTAL OF ABOVE CALCULATIONS				=	\$ 860
Reduction of ½ for filing by small entity, if applicable. A Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).				\$ 0	
SUBTOTAL				=	\$ 860
Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).				\$ 0	
TOTAL NATIONAL FEE				=	\$ 860
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property				\$ 0	
TOTAL FEES ENCLOSED				=	\$ 860
Amount to be refunded:				\$ -----	
charged:				\$ 860.00	
<p>a. <input type="checkbox"/> A check in the amount of \$ _____ to cover the above fees is enclosed.</p> <p>b. ■ Please charge my Deposit Account No. <u>01-1250</u> in the amount of \$ <u>860.00</u> to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. <u>01-0243</u>.</p> <p>c. ■ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>01-1250</u>. A duplicate copy of this sheet is enclosed.</p> <p>NOTE: When an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.</p>					
SEND ALL CORRESPONDENCE TO:		Henkel Corporation, Law Dept. 2500 Renaissance Blvd., Suite 200 Gulph Mills, PA 19406			
		SIGNATURE: <u>Kimberly R. Hild</u>			
		Kimberly R. Hild NAME ATTORNEY FOR APPLICANT 39,224 REGISTRATION NUMBER			

Express Mail Label No. EL615774771US

PATENT
Docket H 3381 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Re: PCT/EP99/06799

International Filing Date: September 14, 1999
Priority Date Claimed: September 21, 1998
Applicant: Kolowrot et al.
Title: A SPRAYABLE HOTMELT ADHESIVE
Applicants' Reference: H 3381 PCT/US

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Box PCT
Washington, DC 20231

Attn: DO/EO/US

Sir:

Before examining this application, please enter these amendments:

IN THE CLAIMS:

Please cancel claims 2 to 10 without prejudice.

Should any fees be deemed necessary to enter this amendment, please charge them to Deposit
Account No. 01-1250.

Respectfully submitted,

Kimberly R. Hild
Kimberly R. Hild
Reg. No. 39,224
Attorney for Applicants
(610) 278-4964

Date: March 16, 2001
Henkel Corporation Patent Department
2500 Renaissance Blvd., Suite 200
Gulph Mills, PA 19406
KRH/bj

09787248-082401

VERSION WITH MARKINGS TO SHOW CHANGES MADE

In the Specification:

On page 6, the paragraph beginning on line 3 and ending on line 5 has been amended as follows:

The poly- α -olefin mixture preferably contains at least one poly- α -olefin with a melt viscosity of 40,000 to 60,000 and at least one poly- α -olefin with a melt viscosity of 3,000 to 10,000 [Pas] mPas at 190°C.

On page 17, line 1, the heading "CLAIMS" has been amended as shown below:
[CLAIMS] What is claimed is:

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PATENT
Docket No. H 3381 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:
Kolowrot et al.,

International Application No. PCT/EP99/06799
International Filing Date: September 14, 1999

Serial No. 09/787,248 **Examiner:** To be assigned
Filed: To be assigned **Art Unit:** To be assigned

Title: SPRAYABLE HOT-MELT ADHESIVE

"Express Mail Post Office to Addressee" service mailing label number EL615774900US

SUPPLEMENTAL PRELIMINARY AMENDMENT

Box PCT
Assistant Commissioner for Patents
Washington, DC 20231

Attn: DO/EO/US

Sir:

Prior to examining this application, please amend the application as follows:

In the Specification (Using the Substitute English Translation):

On page 1 of the English translation, on a separate line between the title and line 1, please insert the following header and paragraph on consecutive lines as shown below:

09787248-082401

-- CROSS REFERENCE TO RELATED APPLICATIONS

This application is a national stage application under 35 U.S.C. § 371 of international application PCT/EP99/06799 filed on September 14, 1999, the international application not being published in English. This application also claims priority under 35 U.S.C. § 119 to DE 198 43 141.4, filed on September 21, 1998. --

On page 1, on a separate line immediately after the above inserted paragraph and before line 1, please insert the following header:

-- FIELD OF THE INVENTION --.

On page 1, on a separate line between lines 5 and 6, please insert the following header:

-- BACKGROUND OF THE INVENTION -- .

On page 3, on a separate line between lines 14 and 15, please insert the following header:

-- SUMMARY OF THE INVENTION --.

On page 4, on a separate line between lines 15 and 16, please insert the following header:

-- DETAILED DESCRIPTION OF THE INVENTION --.

On page 6, please replace the paragraph beginning on line 3 and ending on line 5 with the following new paragraph:

-- The poly- α -olefin mixture preferably contains at least one poly- α -olefin with a melt viscosity of 40,000 to 60,000 and at least one poly- α -olefin with a melt viscosity of 3,000 to 10,000 mPas at 190°C. --

On page 17, line 1, please replace the heading "CLAIMS" with the following heading:

-- What is claimed is: --

On a separate page, after page 19, please insert the enclosed Abstract of the Disclosure.

In the Claims

Please cancel Claim 1, without prejudice.

Please add the following new claims:

-- 15. (NEW) A sprayable hotmelt adhesive comprising:

A) 30 weight percent to 70 weight percent of one or more poly- α -olefins, wherein the poly- α -olefin or the mixture of poly- α -olefins has a softening point of 70°C to 130°C and a melt viscosity at 190°C of 1,000 mPas to 20,000 mPas;

B) 5 weight percent to 30 weight percent of at least one oil; and

C) 20 weight percent to 60 weight percent of at least one hydrocarbon resin having a softening range of 70°C to 140°C;

wherein the hotmelt adhesive has a viscosity of 500 mPas to 4,000 mPas at 150°C.

16. (NEW) The hotmelt adhesive of claim 15 wherein at least one of the poly- α -olefins has a weight average molecular weight, as determined by gel permeation chromatography, of at most 100,000, or a number average molecular weight, as determined by gel permeation chromatography, of at least 4,000, wherein the difference between the weight average and the number average molecular weight is no more than six times the number average molecular weight.

17. (NEW) The hotmelt adhesive of claim 15 wherein the hotmelt adhesive viscosity ranges from 700 mPas to 1,900 mPas at 150°C, as measured in accordance with ASTM D 3236-88.

18. (NEW) The hotmelt adhesive of claim 15, wherein the poly- α -olefin or the mixture of poly- α -olefins is substantially amorphous and at least one of the poly- α -olefins comprises polymerized units of:

- (i) 3 weight percent to 75 weight percent of an α -olefin containing 4 to 10 carbon atoms,
- (ii) 25 weight percent to 95 weight percent of propene, and
- (iii) 0 to 20 weight percent of ethene.

19. (NEW) The hotmelt adhesive of claim 15, wherein the poly- α -olefin or the mixture of poly- α -olefins has a melt viscosity at 190°C of 2,000 mPas to 15,000 mPas.

20. (NEW) The hotmelt adhesive of claim 15, wherein at least one of the poly- α -olefins has a density of less than 0.9 g/cm³, a needle penetration of 8 mm to 4.0 mm, and a weight average molecular weight, as determined by gel permeation chromatography, of at most 100,000, or a number average molecular weight, as determined by gel permeation chromatography, of at least 4,000, wherein the difference between the weight average and the number average molecular weight is no more than six times the number average molecular weight.

21. (NEW) The hotmelt adhesive of claim 15, wherein the hotmelt adhesive comprises at least one first poly- α -olefin having a melt viscosity of 40,000 mPas to 60,000 mPas, and at least one second poly- α -olefin having a melt viscosity of 3,000 mPas to 10,000 mPas at 190°C.

22. (NEW) The hotmelt adhesive of claim 15, wherein the paraffinic oil comprises a medicinal white oil.

23. (NEW) The hotmelt adhesive of claim 15, wherein the hydrocarbon resin comprises a hydrocarbon resin having 5 to 9 carbon atoms.

24. (NEW) The hotmelt adhesive of claim 15, wherein the hotmelt adhesive further comprises an additive selected from a heat or light stabilizer, an optical brightener, an antistatic agent, a lubricant or antiblocking agent, a nucleating agent, a dye, a pigment or a flame retardant, or combinations thereof.

25. (NEW) The hotmelt adhesive of claim 15, wherein the combined amount of the oil and the hydrocarbon resin is at least 30 weight percent, based on the total weight of the poly- α -olefins, the oil, and the hydrocarbon resin.

26. (NEW) The hotmelt adhesive of claim 15, wherein the poly- α -olefins, the oil, and the hydrocarbon resin are selected so that the mixture of the poly- α -olefins, the oil, and the hydrocarbon resin has a viscosity at 100°C ranging from 5 Pas to 15 Pas, wherein the viscosity may vary plus or minus 15% from the viscosity range and wherein the viscosity is measured at a shear rate ranging from 2 sec⁻¹ to 250 sec⁻¹.

27. (NEW) A method of bonding sanitary products comprising applying the hotmelt adhesive of claim 15 to a sanitary product.

28. (NEW) The method of claim 27, wherein at least one of the poly- α -olefins has a weight average molecular weight, as determined by gel permeation chromatography, of at most 100,000, or a number average molecular weight, as determined by gel permeation chromatography, of at least 4,000, wherein the difference between the weight average and the number average molecular weight is no more than six times the number average molecular weight.

29. (NEW) The method of claim 27 wherein the sanitary product is a diaper, panty liner or a sanitary napkin.

30. (NEW) A method of bonding films comprising applying to a film the hotmelt adhesive of claim 15 at an application temperature ranging from 120°C to 180°C and at an application weight ranging from 2 g/m² to 10 g/m².

31. (NEW) The method of claim 30 wherein at least one of the poly- α -olefins has a weight average molecular weight, as determined by gel permeation chromatography, of at most 100,000, or a number average molecular weight, as determined by gel permeation chromatography, of at least 4,000, wherein the difference between the weight average and the number average molecular weight is no more than six times the number average molecular weight.

32. (NEW) The method of claim 30 wherein the hotmelt adhesive is applied at a rate of 50 m/min to 400 m/min, the application temperature of the adhesive ranges from 140°C to 160°C, and the application weight ranges from 3 g/m² and 4 g/m².

33. (NEW) The method of claim 30 wherein the fiber is a polyolefin, a nonwoven, or combinations thereof.

34. (NEW) A process for preparing a hotmelt adhesive comprising
- A) mixing raw materials comprising
- i) 30 weight percent to 70 weight percent of one or more poly- α -olefins, wherein the poly- α -olefin or the mixture of poly- α -olefins has a softening point of 70°C to 130°C and a melt viscosity at 190°C of 1,000 mPas to 20,000 mPas;
 - ii) 5 weight percent to 30 weight percent of at least one oil; and
 - iii) 20 weight percent to 60 weight percent of at least one hydrocarbon resin having a softening range of 70°C to 140°C; wherein the mixing is performed at a temperature ranging from 150 to 200°C and in an inert gas atmosphere, or in a vacuum, or combinations thereof; and
- B) forming a hotmelt adhesive having a viscosity of 500 mPas to 4,000 mPas at 150°C.
35. (NEW) The process of claim 34 wherein at least one of the poly- α -olefins has a weight average molecular weight, as determined by gel permeation chromatography, of at most 100,000, or a number average molecular weight, as determined by gel permeation chromatography, of at least 4,000, wherein the difference between the weight average and the number average molecular weight is no more than six times the number average molecular weight. --

REMARKS

Applicants respectfully request the Examiner to enter the above amendments prior to the examination of this application. It is also respectfully requested that the English translation being submitted herewith and labeled in the header "Substitute Translation", be substituted for the translation previously submitted, as the previous translation is not a correct translation of PCT/99/06799. This error was noticed after Applicants' first submission under 35 U.S.C. § 371. Applicants authorize the Assistant Commissioner to charge any fees necessary to have this translation entered into the record.

Status of Claims

Claims 15 to 35 will be pending after entry of the present amendment. Claim 1 is being canceled without prejudice.

Amendment

The specification is being amended to insert section headers and an abstract of the disclosure in accordance with 37 CFR §1.77 to better conform with US patent practice. The specification is also being amended to insert a cross-reference to related applications in accordance with 37 CFR §1.78 and to claim priority to those applications listed therein.

The specification is also being amended on page 6, line 5 to correct a typographical error in viscosity units ("Pas" should be "mPas"). For example, it would be readily apparent to one skilled in the art, that based on the viscosity range provided at page 3, line 21 of the specification for the poly- α -olefin, that the units should be "mPas".

Applicants submit herewith "Version With Markings To Show Changes Made" to show changes made to the specification (not including the addition of headers and canceling or adding of claims).

New Claims 15 to 35 replace original Claims 1 to 14, and are being presented to better conform with US patent practice. These new claims are supported by the specification for

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example as shown in the Table below (cites to the specification are for the **Substitute** English translation):

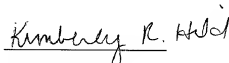
Claim	Support in Specification
15, 16, 17	page 3, line 15 to page 4, line 12
18	page 5, lines 23 to 28
19	page 6, lines 6 to 7
20, 28, 31, 35	page 3, lines 18 to 26
21	page 6, lines 3 to 6
22	page 6, lines 10 to 20
23	page 6, lines 21 to 27
24	page 7, lines 17 to 20
25	page 6, lines 28 to 30
26	page 7, lines 4 to 10, original claim 11
27, 29	page 11, lines 18 to 20, original claim 13
30, 32, 33	page 11, lines 7 to 17
34	page 8, lines 4 to 8

No new matter is added by the new claims or amendments to the specification.

CONCLUSION

The Assistant Commissioner is authorized to charge any deficiency in the required fee or to credit any overpayment to Deposit Account 01-1250 in connection with this amendment.

Respectfully submitted,



Kimberly R. Hild
(Reg. No. 39,224)
Attorney for Applicants
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Henkel Corporation
Law Department
2500 Renaissance Boulevard, Suite 200
Gulph Mills, PA 19406

A Sprayable Hotmelt Adhesive

This invention relates to a sprayable hotmelt adhesive based on a substantially amorphous poly- α -olefin (APAO) with a softening point (ring-and-ball method) of 70 to 130°C, a melt viscosity at 190°C of 1,000 to 20,000 mPas, a density of <0.90 g/cm³, a needle penetration of 8 to 40 0.1 mm and a molecular weight of at most 100,000 (weight average) or at least 4,000 (number average), the difference between the weight average and the number average not exceeding 6 times the number average, with an addition of resins and oils. The present invention also relates to the production of this hotmelt adhesive and to its use.

A sprayable hotmelt adhesive is already known. Thus, EP 0 442 045 B1 describes a sprayable hotmelt adhesive based on the above-mentioned poly- α -olefin with no more than 30% by weight of additives, more especially waxes and/or resins. Paraffinic or aromatic oils may also be used. The Examples contain ethene/propene/1-butene terpolymers and plastic microwaxes and, in some cases, even a hydrocarbon resin. The viscosities are in the range from 2,700 to 3,400 mPas at 190°C.

The Hüls technical information pamphlet entitled "Vestoplast: Klebstoff für Hot-Melt-Anwendungen (Vestoplast: an Adhesive Raw Material for Hotmelt Applications)" dating back to 1996 describes a number of atactic poly- α -olefin copolymers and terpolymers of the monomers ethylene; propylene and 1-butene. Polymers such these have excellent wetting behavior on nonpolar substrates such as, for example, polyethylene and polypropylene. An increase in adhesive strength can be achieved by combination with tackifying resins or plasticizers. Recommended resins include, in particular, substantially saturated hydrocarbon resins. Added in a quantity of up to 30%, the resin acts as a

- plasticizer. Beyond 35%, the mixture assumes the properties of the resin and becomes hard and brittle. The tensile strength of a mixture of poly- α -olefin and resin in a ratio of 70:30 parts by weight falls by at least 20% when the ratio is changed to 50:50 parts by weight at the expense of the poly- α -olefin. Waxes are added inter alia for fine viscosity adjustment and to reduce stringing. In most cases, polybutenes of relatively low molecular weight are recommended as plasticizers. It costs less to use paraffinic or naphthenic mineral oils. The basic formulation for sprayable hotmelt adhesives for sanitary articles contains 70 parts of Vestoplast 704 (a poly- α -olefin), 25 parts of Escorez 5380 (a hydrogenated polycyclopentadiene), 5 parts of Napvis D 10 (a polybutene liquid at room temperature) and 0.4 part of Irganox 1010 (an antioxidant for polyolefins). This known sprayable hotmelt adhesive has a melt viscosity of 2,000 mPas at 190°C and a softening point of 91°C. Unfortunately, it has poor adhesive properties.
- Thus, the initial adhesion values in particular are poor and deteriorate even further in the event of storage. Further disadvantages include a distinct reduction in the adhesion values in the wet state, the so-called "wet values" or "wet strength". Since the adhesives described in the document in question are used in particular for the production of a composite material, for example for a composite material containing at least one nonwoven, it is essential that the adhesion values after storage and the wet adhesion values do not fall below the initial values. The adhesives are being used to an increasing extent in many areas of everyday life because, in general, they represent a simple, permanent and safe method of fixing materials.
- Composite materials of the type used in particular in the field of personal hygiene are generally materials with a limited useful life which are discarded, for example, after being used only once, more especially disposable products. Products such as these mainly include diapers, sanitary towels and panty liners etc. of which the function is to absorb

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bodily fluids such as, for example, urine and blood. In view of this fact, the adhesive used in the composite material must hold the material together even in the "wet" state. This is only possible if the so-called wet values are at the same level as the initial values.

5 Accordingly, the problem addressed by the present invention was to improve adhesion, above all initial adhesion, but also adhesion after storage for a certain period and also wet adhesion. In addition, however, the adhesion levels would be made more independent of the processing conditions, more especially the spraying temperature, the quantity applied
10 and the method of application. However, the spraying properties and other important processing and performance properties would not be adversely affected.

 The solution provided by the invention is defined in the claims and consists essentially in the composition of the sprayable hotmelt adhesive,
15 namely

- A) 30 to 70% by weight and preferably 35 to 50% by weight of at least one substantially amorphous poly- α -olefin with a softening point (ring-and-ball method) of 70 to 130°C, a melt viscosity at 190°C of 1,000 to 20,000 mPas, a density of <0.90 g/cm³, a needle penetration of 8 to 40
20 0.1 nm, a molecular weight of at most 100,000 (weight average) or at least 4,000 (number average), the difference between the weight average and the number average molecular weight being no more than six times the number average,
- B) 5 to 30% by weight and more particularly 15 to 25% by weight of at
25 least one oil of a saturated hydrocarbon of relatively low vapor pressure which is liquid at 20°C, more especially at least one mineral oil with a paraffinic or naphthenic base and, above all, at least one medicinal white oil,
- C) 20 to 60% by weight and more particularly 25 to 50% by weight of at

least one hydrocarbon resin with a softening range of 70 to 140°C and, more particularly, 80 to 120°C, suitable hydrocarbon resins being above all resins containing 5 to 9 carbon atoms and including partly or completely hydrogenated, aliphatic and aromatic hydrocarbon resins, polyterpene resins and modified polyterpene resins and also natural resins, and

D) optionally additives, such as heat and light stabilizers, optical brighteners, antistatic agents, lubricants and antiblocking agents, nucleating agents, dyes, pigments and flame retardants,

with a viscosity of 500 to 4,000 mPas and, more particularly, in the range from 700 to 1,900 mPas, as measured in accordance with ASTM D 3236-88 (Brookfield Model RVT DV II, 150°C, spindle 27).

The poly- α -olefins (component A) are polymers of the type described in EP 0 442 045 B1 of which the disclosure is hereby specifically included as part of the present application.

Preferred polyolefins are either completely amorphous or have only minimal crystallinity. Accordingly, the heat of fusion for heating to 180°C (ΔH) by DTA analysis in accordance with DIN 53765 should only be between 330 and 500 J/g and, more particularly, between 370 and 450 J/g while the specific heat capacity from 20 to 200°C by DTA analysis in accordance with DIN 53765 should be between 1.0 and 4.0 J/g·K and, more particularly, between 1.7 and 3.2 J/g·K. The glass transition temperature T_g , which is also to be determined by DTA analysis in accordance with DIN 53765, should be in the range from -15°C to -40°C and, more particularly, in the range from -20°C to -38°C. Suitable products may be produced, for example, by radical degradation of commercially available substantially amorphous poly- α -olefins with softening points of 80 to 140°C and viscosities of 5,000 to 100,000 mPas at 190°C. This method of production is the subject of German patent application P 40 00 695.6. However, they

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may also be produced by any other process providing they then have the claimed features.

Suitable substantially amorphous poly- α -olefins may carry functional groups to improve adhesion to standard substrates. These functional groups may be introduced either by copolymerization with small quantities of functional monomers or, preferably, by radical reaction of commercially available poly- α -olefins with such functional monomers. Suitable monomers are, for example, maleic anhydride, fumaric acid, acrylic and methacrylic acid, itaconic acid, aconitic acid and derivatives thereof such as, for example, esters or amides and vinyl trimethoxysilane (VTMO) and 3-methacryloxypropyl trimethoxysilane (MEMO; $\text{H}_2\text{C} = \text{C}(\text{CH}_3)\text{COO}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$). They are normally used in quantities of 0.1 to 10% by weight and preferably in quantities of 0.5 to 5% by weight, based on the poly- α -olefin. Standard radical initiators such as, for example, dicumyl peroxide or 2,2'-azo-bis-(2-acetoxyp propane) are used in quantities of 0.05 to 3% by weight and preferably 0.1 to 2% by weight. The grafting reaction then takes place at elevated temperatures, generally in the range from 100 to 300°C. Polymers with high cohesion and improved adhesion to certain substrates, such as metal, plastic or glass surfaces, are obtained in this way.

In one preferred embodiment, the substantially amorphous poly- α -olefin is a binary or ternary copolymer of olefins containing 2 to 10 carbon atoms. This copolymer preferably has the following monomer composition: 3 to 75% by weight of an α -olefin containing 3 to 10 carbon atoms, 25 to 95% by weight of propene and 0 to 20% by weight of ethene.

In addition, the copolymer may also contain functional monomers, as already mentioned. In one particularly preferred embodiment, 1-butene is used as the α -olefin containing 3 to 10 carbon atoms. Mixtures of

different poly- α -olefins according to the invention may of course also be used. Poly- α -olefins such as these are commercially available under such names as Vestoplast®, Rexene®, etc.

Component B comprises oils of substantially saturated hydrocarbons of relatively low vapor pressure which are liquid and, in particular, thinly liquid (20 to 300 mPas) at 20°C, more especially mineral oils with either a paraffinic base or a naphthenic base and, above all, medicinal white oils which are approved for use in foods according to FDA 175.105. These white oils have a viscosity of 25 to 230 mPas and, more particularly, in the range from 110 to 230 mPas, as measured in accordance with DIN 51562 at 20°C. These oils are used inter alia for fine viscosity adjustment. They are commercially available under such names as Primol352®, Essomarc® oils, Pionier 0352®, Drakeol 35®, Kaydol®, Ondina G® oils, Catenex N® oils, etc.

Component C contains a substantially and, more particularly, completely hydrogenated polycyclopentadiene resin as the hydrocarbon resin. Specific examples are MBG® resins, Regalite R and S® resins, Zonarez® resins, Zonatac® resins, Betaprene® AC, AF, AK, AR, B, BC, BR resins, Betalite® resins, Eastotac® resins, ECR® resins, Escorez® resins, Wingtack® resins, etc.

Components B and C together should make up at least 30% by weight, preferably at least 35% by weight and more preferably at least 45% by weight, based on the sum of components A, B and C. In other words, the poly- α -olefin content should be at most 70%, more particularly at most 65% and above all at most 55% by weight, based on components A, B and C.

Besides these essential components, other substances, for example heat and light stabilizers, optical brighteners, antistatic agents, lubricants and antiblocking agents, nucleating agents and dyes, pigments and flame

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retardants, may also be added.

A suitable antioxidant is, above all, Irganox 1010 used in a quantity of 0 to 3% by weight, based on the hotmelt adhesive as a whole. Other suitable stabilizers are, above all, Irganox PS 800, Irgastab DBTM, Tinuvin P, Wingstay stabilizers, Wingstay SN-1, Evernox 10, etc.

Fully synthetic thermoplastic polymers, especially polypropylene, may also be added, for example to modify the softening temperature. Suitable polypropylene types include atactic polypropylene, homopolypropylene, statistical copolymers, block copolymers and graft copolymers of propylene. However, other synthetic polymers may also be used, including for example ethylene/vinyl acetate copolymers, ethylene/acrylic acid copolymers, polyisobutene, polybutene, isotactic poly-1-butene, styrene block copolymers and rubber. They make up from 0 to 50% by weight and preferably from 15 to 45% by weight of the hotmelt adhesive as a whole.

To produce the hotmelt adhesive, the raw materials mentioned above are mixed in the melt in an inert gas atmosphere and/or in a vacuum until they are homogeneous. The temperature of the melt is preferably from 150 to 200°C. It is important to ensure that there are no specks in the hotmelt adhesive produced.

The hotmelt adhesive obtained is largely characterized by its viscosity of 500 to 4,000 mPas and preferably 700 to 1,900 mPas at 150°C, as measured in accordance with ASTM D 3236-88 (Brookfield Model RVT DVII; 150°C; spindle 27). It is also characterized by its favorable sprayability both in regard to "atomization" of the melt and in regard to so-called "spin spraying". In the first of these two processes, the melt is atomized into fine melt droplets whereas, in "spin spraying", a melt strand leaves the spray nozzle in a spiral and, without breaking, is deposited in a spiral pattern onto the substrate to be coated. This spraying technology generally allows contactless application and the coating of uneven irregular surfaces. It is also suitable for applications where, although bonding over a

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certain area may be required, the bonded area must be permeable to air or moisture. The fact that the adhesive is not applied over the entire area means that material can be saved.

Another advantage arises in the coating of heat-sensitive substrates.

- 5 By virtue of the contactless application and the very low heat content of the melt or melt strand applied, the substrate undergoes less stressing or damage than is the case in conventional methods of application. Hotmelt spraying technology is environmentally friendly and, overall, covers many applications which, hitherto, have been the preserve of sprayed solvent-based adhesives.

- 10 The favorable sprayability naturally applies both to the "atomization" mentioned earlier on and to "spin spraying". A uniform spray pattern produced by spin spraying requires a viscosity of the adhesive to be sprayed which is largely independent of the shear rate at the particular application temperature. If this were not the case, it would not be possible to achieve a constant spraying width, for example in intermittent spraying. Both at the beginning and at the end of the particular spraying cycle, the viscosity would be increased on account of the lower shear rate so that the spray pattern would change.

- 20 The spraying behavior is distinguished in particular by the following properties:

- a) Temperature variations between 140 and 160°C affect the adhesion values to an extent of only $\pm 14\%$, based on a coating weight of 4 g/m².
b) Variations in the coating weight of 2 to 4 g/m² affect the adhesion values to an extent of only $\pm 7\%$.
25 c) Such methods of application as "Control Coat" or spin spraying also have only a slight effect (namely $\pm 6\%$) on the adhesion values.

- Other positive properties of the hotmelt adhesive according to the invention include not only its high early adhesion, but also the increase in
30 the adhesion values after ageing. They are in the range from 20 to 60%.

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Other positive properties of the adhesives according to the invention include the increase in wet strength by 29% to 126% and fiber failure of the composite material after ageing both in regard to wet strength and in regard to early adhesion.

- 5 The hotmelt adhesives according to the invention are suitable for bonding various materials. Thus, such materials as polyolefin films, for example polyethylene films or polypropylene films, polyolefin nonwovens, for example polyethylene nonwovens or polypropylene nonwovens, polyurethane films, polyurethane foams, films or moldings of cellulose derivatives, for example tissues, films or moldings of polyacrylates or polymethacrylates, films or moldings of polyesters, can be bonded to one another. The hotmelt adhesives according to the invention may be used to bond materials of the same kind and materials of different kinds to one another.
- 10 In one preferred embodiment of the process according to the invention, the adhesive is used in the production of a composite material containing at least one nonwoven layer. In a particularly preferred embodiment, the nonwoven layer is a polypropylene spunbonded with a weight per unit area of about 10 to about 30 g/m² and, more particularly, about 15 to about 20 g/m².
- 15 A nonwoven layer, preferably one containing or consisting of a polyolefin, is particularly suitable as the second material to be bonded to the substrate.
- 20 In the context of the present invention, a "nonwoven layer" is understood to be a flexible material which is not produced by conventional warp/weft weaving or by loop formation, but rather by the interlocking and/or cohesive and/or adhesive bonding of textile fibers. Accordingly, nonwovens are understood to be loose materials of spun fibers or filaments, generally of polypropylene, polyester or viscose, which are
- 25 generally held together by the adhesion inherent in the fibers. The
- 30

individual fibers may have a preferential orientation (oriented or cross-laid nonwovens) or no orientation (random laid nonwovens). The nonwovens may be mechanically strengthened by needle punching, stitching or lacing by sharp jets of water (so-called spunlaced nonwovens). Adhesively strengthened nonwovens are formed by bonding the fibers with liquid binders (for example acrylate polymers, SBR/NBR, polyvinyl ester or polyurethane dispersions) or by melting or dissolving so-called binding fibers which have been added to the nonwoven during its production. In the case of cohesive strengthening, the fiber surfaces are dissolved by suitable chemicals and joined by pressure or welded at elevated temperature. Nonwovens of so-called spunbondeds, i.e. materials produced by spinning and subsequent deposition, blowing or floating on a conveyor belt, are known as spunbonded nonwovens. Nonwovens containing additional filaments, woven or knitted fabrics count as reinforced or strengthened nonwovens.

In one preferred embodiment of the process according to the invention, the adhesive is used for the production of a composite material containing at least one nonwoven layer. In a particularly preferred embodiment, the nonwoven layer is a polypropylene spunbonded with a weight per unit area of about 10 to about 30 g/m² and, more particularly, in the range from about 15 to about 20 g/m².

By virtue of these positive properties, the hotmelt adhesives according to the invention are suitable for structural bonding in sanitary products, more especially for bonding diapers and sanitary napkins.

Further applications are in the packaging industry, the automotive supply industry, the building industry, etc.

The invention is illustrated by the following Examples.

I Starting materials for the production of the hotmelt adhesives

1. Poly- α -olefins: Vestoplast 508, 704 and 750 (terpolymers of ethylene, propylene and butylene) of Hüls AG
2. Oils: Primol 352, a medicinal white oil manufactured by Esso
- 5 3. Resins: MGB-278 and Regalite R-R91 (cyclopentadiene resins) of Hercules
4. Antioxidants: Irganox 1010 of Ciba Geigy.

II Production and properties of the hotmelt adhesives

- 10 To produce the hotmelt adhesives, the starting materials mentioned above are melted at 160 to 170°C in the quantities shown in Table 1a) in the following order: oil + antioxidant, resin and poly- α -olefin, and stirred at 160 to 170°C until the mixture is speck-free (homogeneous). This generally takes 2 to 4 hours. It is also advisable to apply a vacuum and/or
- 15 an inert gas atmosphere (nitrogen).

The hotmelt adhesives obtained have the properties set out in Table 1b).

III Adhesive testing of the hotmelt adhesives

- 20 1. The spraying tests were carried out with a Meltex CT 325 sprayer (manufacturer: Meltex, Lüneburg). The spray heads used were Nordson models CF 203 and CF 205 (the abbreviation CF stands for controlled fiberization). In the CF spray heads used here, a spiral movement is imparted to the adhesive issuing from the nozzle by compressed air. This
- 25 procedure provides for the controlled, surface-covering and edge-sharp application of hotmelt adhesives.

Quantity applied:	3.0 to 4.0 g · m ⁻²
Application rate:	100 m · min. ⁻¹
Temperature in the melting tank:	160°C

Temperature of the spraying air: 180°C

Adhesive temperature: 160°C.

- Composite materials were produced on the basis of these parameters. On the one hand, the adhesive was sprayed onto the film while the nonwoven moved in the opposite direction; on the other hand, the adhesive was sprayed onto the nonwoven while the film moved in the opposite direction. In addition to these tests carried out at an adhesive temperature of 160°C (tests A), tests were also carried out at an adhesive temperature of 140°C (tests B).
- The film used was a 4P PE film, i.e. a polyethylene film with a thickness of about 10 to about 50 μm and, more particularly, about 20 to about 30 μm . The nonwoven used was Corosoft Plus H, a nonwoven of polypropylene with a weight per unit area of 14 to 30 g/m^2 and, more particularly, 17 $\text{g} \cdot \text{m}^{-2}$. In case a), the hotmelt adhesive was sprayed onto the film which was then bonded to the nonwoven. In case b), the adhesive was sprayed onto the nonwoven which was then bonded to the film.

2. The adhesion value tests are summarized in Table 1c).

IV Results

By comparison with the reference product, the tests of Examples 1 and 2 show

- 5 • a distinct increase in early strength up to fiber failure irrespective of the coating weight and irrespective of the production of the composite (in other words, there was a distinct improvement in the adhesion values in the film/nonwoven composite and in the nonwoven/film composite),
- a distinct increase in the adhesion values with fiber failure after ageing, based on the early strength, irrespective of the production of the
10 composite and
- a distinct increase in the wet adhesion values with fiber failure, based on the early strength, irrespective of the production of the composite and
- a distinct step-by-step reduction in the adhesion values of the reference
15 product after ageing and in the wet test.

V Description of the test methods:

- Melt viscosity according to ASTM D 3236-88, Brookfield Model RVT DV II, 150°C, spindle 27,
20 • softening point according to ASTM E 28,
- needle penetration (100/25/5) in accordance with DIN 52010 in 0.1 mm,
- peel strength (so-called adhesion value) in accordance with DIN 53530 using a Zwick 1435 tensile tester at 20°C/50% relative air humidity. The 50 mm wide sample is separated at a rate of 300 mm/min. at an angle
25 of 180°. The result is expressed in Ncm^{-1} . The coating of the sample was measured at the earliest after 48 h.
- Ageing behavior was simulated by so-called accelerated ageing (72 h at 60°C with a subsequent cooling time of at least 24 h).
- Wet strength (the sample to be tested is placed for 1 h in a 0.9%

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sodium chloride solution and then tested, internal test).

- Crystallinity by DTA analysis in accordance with DIN 53765.

Table 1a):

Composition of the hotmelt adhesives

Raw materials			Hotmelt adhesive		
Type	Commercial name/percentage		Example 1	Example 2	Comparison
APAO	Vestoplast 704	%	22.5%	35.0%	70%
APAO	Vestoplast 508	%	22.5%	-	-
APAO	Vestoplast 750	%	-	5.0%	-
Plasticizer	Napvis D-10	%	-	-	5.0%
Oil	Primol 352	%	19.0%	19.0%	-
Resin	MBG-278	%	35.0%	20.0%	-
Resin	Regalite (R)-R 91	%	-	20.0%	-
Resin	Escorez	%	-	-	25.0
Antioxidant	Irganox 1010	%	1.0%	1.0%	0.4%

Table 1b):

Properties of the hotmelt adhesives

Property	Example 1	Example 2	Comparison
Melt viscosity [mPas] at 150°C	1485	1137	4580
Needle penetration [(100/25/5) 0.1 mm]	42	89	17
Softening point [°C]	79	76	96

Table 1c):

Adhesion values of the film/nonwoven bonds

a) Adhesive sprayed onto film

Adhesion values	Example 1 A	Example 2 A	Comparison Example A	Example 1 B
Coating in g m^{-2}	ca. 3.6	ca. 3.4	4.0	ca. 4.0
Adhesion value in N cm^{-1}	0.54	0.48	0.28	0.68
Adhesion value in N cm^{-1} after ageing	0.84	0.64	0.20	0.78
Wet strength $\text{N} \cdot \text{cm}^{-1}$	1.22	0.62	0.12	0.80

b) Adhesive sprayed onto nonwoven

Adhesion values	Example 1 A	Example 2 A	Comparison Example A	Example 1 B
Coating in g m^{-2}	3.6	ca. 3.4	3.8	ca. 3.6
Adhesion value in N cm^{-1}	0.70	0.52	0.30	0.70
Adhesion value in N cm^{-1} after ageing	0.84	0.63	0.26	0.62
Wet strength $\text{N} \cdot \text{cm}^{-1}$	1.28	0.62	0.08	0.54

CLAIMS

1. A sprayable hotmelt adhesive with a viscosity of 500 to 4,000 mPas at 150°C, characterized by the following composition:

5 A) 30 to 70% by weight of at least one poly- α -olefin with a softening point (ring-and-ball method) of 70 to 130°C, a melt viscosity at 190°C of 1,000 to 20,000 mPas, a density of $<0.90 \text{ g/cm}^3$, a needle penetration of 8 to 40 0.1 nm, a molecular weight of at most 100,000 (weight average) or at least 4,000 (number average), the difference between the weight average and the number average being no more than six times the number average,

10 B) 5 to 30% by weight of at least one oil,

C) 20 to 60% by weight of at least one hydrocarbon resin with a softening range of 70 to 140°C and, and

D) optionally additives.

15 2. A hotmelt adhesive as claimed in claim 1, characterized by a viscosity of 700 to 1,900 mPas at 150°C, as measured in accordance with ASTM D 3236-88.

3. A hotmelt adhesive as claimed in claim 1, characterized in that it is substantially amorphous and has the following monomer composition:

- 3 to 75% by weight of an α -olefin containing 4 to 10 carbon atoms,
- 25 to 95% by weight of propene and
- 0 to 20% by weight of ethene.

4. A hotmelt adhesive as claimed in claim 1, characterized in that the paraffinic oil is a medicinal white oil.

5. A hotmelt adhesive as claimed in claim 1, characterized in that the hydrocarbon resin is a hydrocarbon resin containing 5 to 9 carbon atoms.

6. A hotmelt adhesive as claimed in claim 1, characterized in that the

additive is at least one substance of the following group: heat and light stabilizer, optical brightener, antistatic agent, lubricant and antiblocking agent, nucleating agent, dye, pigment or flame retardant.

7. A hotmelt adhesive as claimed in claim 1, characterized in that
5 components B and C together make up at least 30% by weight, preferably at least 35% by weight and more preferably at least 45% by weight of the sum of components A+B+C.

8. A process for the production of the hotmelt adhesive claimed in at least one of claims 1 to 7, characterized in that the raw materials are mixed
10 in an inert gas atmosphere and/or in a vacuum at temperatures of 150 to 200°C.

9. The use of the hotmelt adhesive claimed in at least one of claims 1 to 7 for structural bonding in sanitary products, more especially for bonding
diapers, panty liners and sanitary napkins.

10. The use of the hotmelt adhesive claimed in at least one of claims 1 to 7 for bonding films, more particularly of polyolefins, and nonwovens, more particularly of polypropylene, the application temperature being
15 between 120 and 180°C and preferably between 140 and 160°C, the coating weight being between 2 and 10 and preferably between 3 and 4
20 g/m² and the application rate preferably being between 50 and 400 m/min.

Sprayable Hotmelt Adhesive

This invention relates to a sprayable hotmelt adhesive based on a substantially amorphous poly- α -olefin (APAO) with a softening point (ring-and-ball method) of 70 to 130°C, a melt viscosity at 190°C of 1,000 to 20,000 mPas and an addition of resins and oils. The present invention also
5 relates to the production of this hotmelt adhesive and to its use.

A sprayable hotmelt adhesive is already known. Thus, EP 0 442 045 B1 describes a sprayable hotmelt adhesive based on the above-mentioned poly- α -olefin with no more than 30% by weight of additives, more especially waxes and/or resins. Paraffinic or aromatic oils may also
10 be used. The Examples contain ethene/propene/1-butene terpolymers and plastic microwaxes and, in some cases, even a hydrocarbon resin. The viscosities are in the range from 2,700 to 3,400 mPas at 190°C.

The Hüls technical information pamphlet entitled "Vestoplast: Klebstoff für Hot-Melt-Anwendungen (Vestoplast: an Adhesive Raw
15 Material for Hotmelt Applications)" dating back to 1996 describes a number of atactic poly- α -olefin copolymers and terpolymers of the monomers ethylene; propylene and 1-butene. Polymers such these have excellent wetting behaviour on nonpolar substrates such as, for example, polyethylene and polypropylene. An increase in adhesive strength can be
20 achieved by combination with tackifying resins or plasticizers. Recommended resins include, in particular, substantially saturated hydrocarbon resins. Added in a quantity of up to 30%, the resin acts as a plasticizer. Beyond 35%, the mixture assumes the properties of the resin and becomes hard and brittle. The tensile strength of a mixture of poly- α -
25 olefin and resin in a ratio of 70:30 parts by weight falls by at least 20% when the ratio is changed to 50:50 parts by weight at the expense of the poly- α -olefin. Waxes are added inter alia for fine viscosity adjustment and

- to reduce stringing. In most cases, polybutenes of relatively low molecular weight are recommended as plasticizers. It costs less to use paraffinic or naphthenic mineral oils. The basic formulation for sprayable hotmelt adhesives for sanitary articles contains 70 parts of Vestoplast 704 (a poly- α -olefin), 25 parts of Escorez 5380 (a hydrogenated polycyclopentadiene), 5 parts of Napvis D 10 (a polybutene liquid at room temperature) and 0.4 part of Irganox 1010 (an antioxidant for polyolefins). This known sprayable hotmelt adhesive has a melt viscosity of 2,000 mPas at 190°C and a softening point of 91°C. Unfortunately, it has poor adhesive properties.
- Thus, the initial adhesion values in particular are poor and deteriorate even further in the event of storage. Further disadvantages include a distinct reduction in the adhesion values in the wet state, the so-called "wet values" or "wet strength". Since the adhesives described in the document in question are used in particular for the production of a composite material, for example for a composite material containing at least one nonwoven, it is essential that the adhesion values after storage and the wet adhesion values do not fall below the initial values. The adhesives are being used to an increasing extent in many areas of everyday life because, in general, they represent a simple, permanent and safe method of fixing materials.
- Composite materials of the type used in particular in the field of personal hygiene are generally materials with a limited useful life which are discarded, for example, after being used only once, more especially disposable products. Products such as these mainly include diapers, sanitary towels and panty liners etc. of which the function is to absorb bodily fluids such as, for example, urine and blood. In view of this fact, the adhesive used in the composite material must hold the material together even in the "wet" state. This is only possible if the so-called wet values are at the same level as the initial values.

- EP 211 311 describes a self-adhesive absorbent shaped body and a process for the production of a pressure-sensitive hotmelt adhesive

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5 suitable therefor. The pressure-sensitive hotmelt adhesive consists of 20 to 80% of substantially amorphous olefinic polymers, 20 to 80% of tackifying resins and 0 to 80% of plasticizer oil, the polymers being atactic and the pressure-sensitive hotmelt adhesive having a shear strength of less than 1 minutes (standard test against steel). Only one olefinic polymer is mentioned. It has a melt viscosity at 190°C of ca. 50,000 mPas.

10 Accordingly, the problem addressed by the present invention was to improve adhesion, above all initial adhesion, but also adhesion after storage for a certain period and also wet adhesion. In addition, however, the adhesion levels would be made more independent of the processing conditions, more especially the spraying temperature, the quantity applied and the method of application. However, the spraying properties and other important processing and performance properties would not be adversely affected.

15 The solution provided by the invention is defined in the claims and consists essentially in the composition of the sprayable hotmelt adhesive, namely

- 20 A) 30 to 70% by weight and preferably 35 to 50% by weight of at least one substantially amorphous poly- α -olefin or poly- α -olefin mixture with a softening point (ring-and-ball method) of 70 to 130°C, a melt viscosity at 190°C of 1,000 to 20,000 mPas and preferably with a density of <0.90 g/cm³, a needle penetration of 8 to 4.0 mm, a molecular weight as determined by gel permeation chromatography of at most 100,000 (weight average) or at least 4,000 (number average), the difference
- 25 between the weight average and the number average molecular weight being no more than six times the number average,
- 30 B) 5 to 30% by weight and more particularly 15 to 25% by weight of at least one oil of a saturated hydrocarbon of relatively low vapor pressure which is liquid at 20°C, more especially at least one mineral oil with a paraffinic or naphthenic base and, above all, at least one medicinal

white oil,

- 5 C) 20 to 60% by weight and more particularly 25 to 50% by weight of at least one hydrocarbon resin with a softening range of 70 to 140°C and, more particularly, 80 to 120°C, suitable hydrocarbon resins being above all resins containing 5 to 9 carbon atoms and including partly or completely hydrogenated, aliphatic and aromatic hydrocarbon resins, polyterpene resins and modified polyterpene resins and also natural resins, and
- 10 D) optionally additives, such as heat and light stabilizers, optical brighteners, antistatic agents, lubricants and antiblocking agents, nucleating agents, dyes, pigments and flame retardants,

- with a viscosity of 500 to 4,000 mPas and, more particularly, in the range from 700 to 1,900 mPas, as measured in accordance with ASTM D 3236-15 88 (Brookfield Model RVT DV II, 150°C, spindle 27).

The poly- α -olefins (component A) are polymers of the type described in EP 0 442 045 B1 of which the disclosure is hereby specifically included as part of the present application.

- 20 Preferred polyolefins are either completely amorphous or have only minimal crystallinity. Accordingly, the heat of fusion for heating to 180°C (ΔH) by DTA analysis in accordance with DIN 53765 should only be between 330 and 500 J/g and, more particularly, between 370 and 450 J/g while the specific heat capacity from 20 to 200°C by DTA analysis in accordance with DIN 53765 should be between 1.0 and 4.0 J/g·K and, more particularly, between 1.7 and 3.2 J/g·K. The glass transition temperature T_g , which is also to be determined by DTA analysis in accordance with DIN 53765, should be in the range from -15°C to -40°C and, more particularly, in the range from -20°C to -38°C. Suitable products may be produced, for example, by radical degradation of commercially available substantially
- 25
- 30 amorphous poly- α -olefins with softening points of 80 to 140°C and

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viscosities of 5,000 to 100,000 mPas at 190°C. This method of production is the subject of German patent application P 40 00 695.6. However, they may also be produced by any other process providing they then have the claimed features.

- 5 Suitable substantially amorphous poly- α -olefins may carry functional groups to improve adhesion to standard substrates. These functional groups may be introduced either by copolymerization with small quantities of functional monomers or, preferably, by radical reaction of commercially available poly- α -olefins with such functional monomers. Suitable mono-
- 10 mers are, for example, maleic anhydride, fumaric acid, acrylic and methacrylic acid, itaconic acid, aconitic acid and derivatives thereof such as, for example, esters or amides and vinyl trimethoxysilane (VTMO) and 3-methacryloxypropyl trimethoxysilane (MEMO; $\text{H}_2\text{C} = \text{C}(\text{CH}_3)\text{COO}(\text{CH}_2)_3\text{Si}(\text{OCH}_3)_3$). They are normally used in quantities of 0.1 to 10% by weight
- 15 and preferably in quantities of 0.5 to 5% by weight, based on the poly- α -olefin. Standard radical initiators such as, for example, dicumyl peroxide or 2,2'-azo-bis-(2-acetoxyp propane) are used in quantities of 0.05 to 3% by weight and preferably 0.1 to 2% by weight. The grafting reaction then takes place at elevated temperatures, generally in the range from 100 to
- 20 300°C. Polymers with high cohesion and improved adhesion to certain substrates, such as metal, plastic or glass surfaces, are obtained in this way.

- In one preferred embodiment, the substantially amorphous poly- α -olefin is a binary or ternary copolymer of olefins containing 2 to 10 carbon
- 25 atoms. This copolymer preferably has the following monomer composition: 3 to 75% by weight of an α -olefin containing 3 to 10 carbon atoms, 25 to 95% by weight of propene and 0 to 20% by weight of ethene.

- In addition, the copolymer may also contain functional monomers, as
- 30 already mentioned. In one particularly preferred embodiment, 1-butene is

used as the α -olefin containing 4 to 10 carbon atoms. Mixtures of different poly- α -olefins according to the invention may of course also be used.

The poly- α -olefin mixture preferably contains at least one poly- α -olefin with a melt viscosity of 40,000 to 60,000 and at least one poly- α -olefin with a melt viscosity of 3,000 to 10,000 Pas at 190°C.

The melt viscosity of the poly- α -olefin or the poly- α -olefin mixture is advantageously in the range from 2,000 to 15,000 mPas at 190°C.

Poly- α -olefins such as these are commercially available under such names as Vestoplast®, Rexene®, etc.

10 Component B comprises oils of substantially saturated hydrocarbons of relatively low vapor pressure which are liquid and, in particular, thinly liquid (20 to 300 mPas) at 20°C, more especially mineral oils with either a paraffinic base or a naphthenic base and, above all, medicinal white oils which are approved for use in foods under FDA 175 105. These white oils
15 have a viscosity of 25 to 230 mPas and, more particularly, in the range from 110 to 230 mPas, as measured in accordance with DIN 51562 at 20°C. These oils are used inter alia for fine viscosity adjustment. They are commercially available under such names as Primol352®, Essomarc® oils, Pionier 0352®, Drakeol 35®, Kaydol®, Ondina G® oils, Catenex N® oils, etc.

20 Component C contains above all a hydrocarbon resin of C₅₋₉ structural units, more particularly a substantially and, more particularly, completely hydrogenated polycyclopentadiene resin as the hydrocarbon resin. Specific examples are MBG® resins, Regalite R and S® resins,
25 Zonarez® resins, Zonatac® resins, Betaprene® AC, AF, AK, AR, B, BC, BR resins, Betalite® resins, Eastotac® resins, ECR® resins, Escorez® resins, Wingtack® resins, etc.

Components B and C together should make up at least 30% by weight, preferably at least 35% by weight and more preferably at least 45%
30 by weight, based on the sum of components A, B and C. In other words,

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the poly- α -olefin content should be at most 70%, more particularly at most 65% and above all at most 55% by weight, based on components A, B and C.

In order to obtain a particularly uniform spray pattern, even at relatively low melting temperatures of, for example, 130°C, components A, B and C should be selected within the claimed range so that the melt viscosity at 100°C is in the range from 5 to 15 Pas \pm 15, more particularly \pm 10%, depending on the shear rate between 0 and 250 sec⁻¹. Even at 130°C, it shows non-newtonian flow behaviour. The measurements were carried out to DIN 53018-1.

The hotmelt adhesives according to the invention are preferably non-pressure-sensitive hotmelt adhesives. Besides the percentage contents of components A, B and C, the choice of the nature of the components is very important. Thus, the melt viscosity of the poly- α -olefin or the poly- α -olefin mixture should be between 2 and 15 Pas at 190°C (Brookfield).

Besides these essential components, other substances, for example heat and light stabilizers, optical brighteners, antistatic agents, lubricants and antiblocking agents, nucleating agents and dyes, pigments and flame retardants, may also be added.

A suitable antioxidant is, above all, Irganox 1010 used in a quantity of 0 to 3% by weight, based on the hotmelt adhesive as a whole. Other suitable stabilizers are, above all, Irganox PS 800, Irgastab DBTM, Tinuvin P, Wingstay stabilizers, Wingstay SN-1, Evernox 10, etc.

Fully synthetic thermoplastic polymers, especially polypropylene, may also be added, for example to modify the softening temperature. Suitable polypropylene types include atactic polypropylene, homopolypropylene, statistical copolymers, block copolymers and graft copolymers of propylene. However, other synthetic polymers may also be used, including for example ethylene/vinyl acetate copolymers, ethylene/acrylic acid

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copolymers, polyisobutene, polybutene, isotactic poly-1-butene, styrene block copolymers and rubber. They make up from 0 to 50% by weight and preferably from 15 to 45% by weight of the hotmelt adhesive as a whole.

- 5 To produce the hotmelt adhesive, the raw materials mentioned above are mixed in the melt in an inert gas atmosphere and/or in a vacuum until they are homogeneous. The temperature of the melt is preferably from 150 to 200°C. It is important to ensure that there are no specks in the hotmelt adhesive produced.

- 10 The hotmelt adhesive obtained is largely characterized by its viscosity of 500 to 4,000 mPas and preferably 700 to 1,900 mPas at 150°C, as measured to ASTM D 3236-88 (Brookfield Model RVT DVII; 150°C; spindle 27). It is also characterized by its favorable sprayability both in regard to "atomization" of the melt and in regard to so-called "spin spraying". In the first of these two processes, the melt is atomized into fine
- 15 melt droplets whereas, in "spin spraying", a melt strand leaves the spray nozzle in a spiral and, without breaking, is deposited in a spiral pattern onto the substrate to the coated. This spraying technology generally allows contactless application and the coating of uneven irregular surfaces. It is also suitable for applications where, although bonding over a certain area
- 20 may be required, the bonded area must be permeable to air or moisture. The fact that the adhesive is not applied over the entire area means that material can be saved.

- Another advantage arises in the coating of heat-sensitive substrates. By virtue of the contactless application and the very low heat content of the
- 25 melt or melt strand applied, the substrate undergoes less stressing or damage than is the case in conventional methods of application. Hotmelt spraying technology is environmentally friendly and, overall, covers many applications which, hitherto, have been the preserve of sprayed solvent-based adhesives.

- 30 The favourable sprayability naturally applies both to the

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5 "atomization" mentioned earlier on and to "spin spraying". A uniform spray pattern produced by spin spraying requires a viscosity of the adhesive to be sprayed which is largely independent of the shear rate at the particular application temperature. If this were not the case, it would not be possible to achieve a constant spraying width, for example in intermittent spraying. Both at the beginning and at the end of the particular spraying cycle, the viscosity would be increased on account of the lower shear rate so that the spray pattern would change.

10 The spraying behavior is distinguished in particular by the following properties:

- 15 a) Temperature variations between 140 and 160°C affect the adhesion values to an extent of only $\pm 14\%$, based on a coating weight of 4 g/m².
b) Variations in the coating weight of 2 to 4 g/m² affect the adhesion values to an extent of only $\pm 7\%$.
c) Such methods of application as "Control Coat" or spin spraying also have only a slight effect (namely $\pm 6\%$) on the adhesion values.

20 Other positive properties of the hotmelt adhesive according to the invention include not only its high early adhesion, but also the increase in the adhesion values after ageing. They are in the range from 20 to 60%. Other positive properties of the adhesives according to the invention include the increase in wet strength by 29% to 126% and fiber failure of the composite material after ageing both in regard to wet strength and in regard to early adhesion.

25 The hotmelt adhesives according to the invention are suitable for bonding various materials. Thus, such materials as polyolefin films, for example polyethylene films or polypropylene films, polyolefin nonwovens, for example polyethylene nonwovens or polypropylene nonwovens, 30 polyurethane films, polyurethane foams, films or mouldings of cellulose

derivatives, for example tissues, films or moldings of polyacrylates or polymethacrylates, films or mouldings of polyesters, can be bonded to one another. The hotmelt adhesives according to the invention may be used to bond materials of the same kind and materials of different kinds to one another.

In one preferred embodiment of the process according to the invention, the adhesive is used in the production of a composite material containing at least one nonwoven layer. In a particularly preferred embodiment, the nonwoven layer is a polypropylene spunbonded with a weight per unit area of about 10 to about 30 g/m² and, more particularly, about 15 to about 20 g/m².

A nonwoven layer, preferably one containing or consisting of a polyolefin, is particularly suitable as the second material to be bonded to the substrate.

In the context of the present invention, a "nonwoven layer" is understood to be a flexible material which is not produced by conventional warp/weft weaving or by loop formation, but rather by the interlocking and/or cohesive and/or adhesive bonding of textile fibers. Accordingly, nonwovens are understood to be loose materials of spun fibers or filaments, generally of polypropylene, polyester or viscose, which are generally held together by the adhesion inherent in the fibers. The individual fibers may have a preferential orientation (oriented or cross-laid nonwovens) or no orientation (random laid nonwovens). The nonwovens may be mechanically strengthened by needle punching, stitching or lacing by sharp jets of water (so-called spunlaced nonwovens). Adhesively strengthened nonwovens are formed by bonding the fibers with liquid binders (for example acrylate polymers, SBR/NBR, polyvinyl ester or polyurethane dispersions) or by melting or dissolving so-called binding fibers which have been added to the nonwoven during its production. In the case of cohesive strengthening, the fiber surfaces are dissolved by

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suitable chemicals and joined by pressure or welded at elevated temperature. Nonwovens of so-called spunbondeds, i.e. materials produced by spinning and subsequent deposition, blowing or floating on a conveyor belt, are known as spunbonded nonwovens. Nonwovens containing additional filaments, woven or knitted fabrics count as reinforced or strengthened nonwovens.

In one preferred embodiment of the process according to the invention, the adhesive is used for the production of a composite material containing at least one nonwoven layer, above all for bonding films, more particularly of polyolefins, and nonwovens, more particularly of polypropylene, the application temperature being between 120 and 180°C and preferably between 140 and 160°C, the coating weight varying between 2 and 10 and preferably between 3 and 4 g/m² and the application rate preferably being between 50 and 400 m/min. In a particularly preferred embodiment, the nonwoven layer is a polypropylene spunbonded with a weight per unit area of about 10 to about 30 g/m² and, more particularly, in the range from about 15 to about 20 g/m².

By virtue of these positive properties, the hotmelt adhesives according to the invention are suitable for structural bonding in sanitary products, more especially for bonding diapers and sanitary napkins.

Further applications are in the packaging industry, the automotive supply industry, the building industry, etc.

The invention is illustrated by the following Examples.

25 I Starting materials for the production of the hotmelt adhesives

1. Poly- α -olefins: Vestoplast 508, 704 and 750 (terpolymers of ethylene, propylene and butylene) of Hüls AG
2. Oils: Primol 352, a medicinal white oil manufactured by Esso
3. Resins: MGB-278 and Regalite R-R91 (cyclopentadiene resins) of Hercules

4. Antioxidants: Irganox 1010 of Ciba Geigy.

II Production and properties of the hotmelt adhesives

- To produce the hotmelt adhesives, the starting materials mentioned
- 5 above are melted at 160 to 170°C in the quantities shown in Table 1a) in the following order: oil + antioxidant, resin and poly- α -olefin, and stirred at 160 to 170°C until the mixture is speck-free (homogeneous). This generally takes 2 to 4 hours. It is also advisable to apply a vacuum and/or an inert gas atmosphere (nitrogen).
- 10 The hotmelt adhesives obtained have the properties set out in Table 1b).

III Adhesive testing of the hotmelt adhesives

1. The spraying tests were carried out with a Meltex CT 325 sprayer
- 15 (manufacturer: Meltex, Lüneburg). The spray heads used were Nordson models CF 203 and CF 205 (the abbreviation CF stands for controlled fiberization). In the CF spray heads used here, a spiral movement is imparted to the adhesive issuing from the nozzle by compressed air. This procedure provides for the controlled, surface-covering and edge-sharp
- 20 application of hotmelt adhesives.

	Quantity applied:	3.0 to 4.0 g · m ⁻²
	Application rate:	100 m · min. ⁻¹
	Temperature in the melting tank:	160°C
25	Temperature of the spraying air:	180°C
	Adhesive temperature:	160°C.

- Composite materials were produced on the basis of these
- parameters. On the one hand, the adhesive was sprayed onto the film
- 30 while the nonwoven moved in the opposite direction; on the other hand, the

adhesive was sprayed onto the nonwoven while the film moved in the opposite direction. In addition to these tests carried out at an adhesive temperature of 160°C (tests A), tests were also carried out at an adhesive temperature of 140°C (tests B).

- 5 The film used was a 4P PE film, i.e. a polyethylene film with a thickness of about 10 to about 50 μm and, more particularly, about 20 to about 30 μm . The nonwoven used was Corosoft Plus H, a nonwoven of polypropylene with a weight per unit area of 14 to 30 g/m^2 and, more particularly, 17 $\text{g} \cdot \text{m}^{-2}$. In case a), the hotmelt adhesive was sprayed onto
- 10 the film which was then bonded to the nonwoven. In case b), the adhesive was sprayed onto the nonwoven which was then bonded to the film.

2. The adhesion value tests are summarized in Table 1c).

15 IV Results

By comparison with the reference product, the tests of Examples 1 and 2 show

- a distinct increase in early strength up to fiber failure irrespective of the coating weight and irrespective of the production of the composite (in
- 20 other words, there was a distinct improvement in the adhesion values in the film/nonwoven composite and in the nonwoven/film composite),
- a distinct increase in the adhesion values with fiber failure after ageing, based on the early strength, irrespective of the production of the composite and
- 25 • a distinct increase in the wet adhesion values with fiber failure, based on the early strength, irrespective of the production of the composite and
- a distinct step-by-step reduction in the adhesion values of the reference product after ageing and in the wet test.

V Description of the test methods:

- Melt viscosity to ASTM D 3236-88, Brookfield Model RVT DV II, 150°C, spindle 27,
- softening point to ASTM E 28,
- 5 • needle penetration (100/25/5) to DIN 52010 in 0.1 mm,
- peel strength (so-called adhesion value) to DIN 53530 using a Zwick 1435 tensile tester at 20°C/50% relative air humidity. The 50 mm wide sample is separated at a rate of 300 mm/min. at an angle of 180°. The result is expressed in Ncm^{-1} . The coating of the sample was measured
- 10 at the earliest after 48 h.
- Ageing behavior was simulated by so-called accelerated ageing (72 h at 60°C with a subsequent cooling time of at least 24 h).
- Wet strength (the sample to be tested is placed for 1 h in a 0.9% sodium chloride solution and then tested, internal test).
- 15 • Crystallinity by DTA analysis to DIN 53765.
- Density to DIN 53479
- Flow behavior to DIN 53018-1.

20 The rheological tests of the hotmelts were carried out with a TA Instruments AR 1000 rotational viscosimeter. To this end, a plate/plate geometry (4 cm diameter, 500 μ interval) was used in the shear-rate-dependent flow test. The adhesive was heated for 5 minutes at the measuring temperature between the plate/plate geometry. The viscosity was then measured continuously for 5 minutes as a function of the shear

25 rate of 0 to 250 s^{-1} or 250 to 0 s^{-1} . In the case of samples with newtonian flow behaviour, viscosity is independent of the shear rate.

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Table 1a):

Composition of the hotmelt adhesives

Raw materials			Hotmelt adhesive		
Type	Commercial name/percentage		Example 1	Example 2	Comparison
APAO	Vestoplast 704	%	22.5%	35.0%	70%
APAO	Vestoplast 508	%	22.5%	-	-
APAO	Vestoplast 750	%	-	5.0%	-
Plasticizer	Napvis D-10	%	-	-	5.0%
Oil	Primol 352	%	19.0%	19.0%	-
Resin	MBG-278	%	35.0%	20.0%	-
Resin	Regalite (R)-R 91	%	-	20.0%	-
Resin	Escorez 5380	%	-	-	25.0
Antioxidant	Irganox 1010	%	1.0%	1.0%	0.4%

Table 1b):

Properties of the hotmelt adhesives

Property	Example 1	Example 2	Comparison
Melt viscosity [mPas] at 150°C	1485	1137	4580
Needle penetration [(100/25/5) 0.1 mm]	42	89	17
Softening point [°C]	79	76	96

Table 1c):

Adhesion values of the film/nonwoven bonds

a) Adhesive sprayed onto film

Adhesion values	Example 1 A	Example 2 A	Comparison Example A	Example 1 B
Coating in g m^{-2}	ca. 3.6	ca. 3.4	4.0	ca. 4.0
Adhesion value in N cm^{-1}	0.54	0.48	0.28	0.68
Adhesion value in N cm^{-1} after ageing	0.84	0.64	0.20	0.78
Wet strength $\text{N} \cdot \text{cm}^{-1}$	1.22	0.62	0.12	0.80

b) Adhesive sprayed onto nonwoven

Adhesion values	Example 1 A	Example 2 A	Comparison Example A	Example 1 B
Coating in g m^{-2}	3.6	ca. 3.4	3.8	ca. 3.6
Adhesion value in N cm^{-1}	0.70	0.52	0.30	0.70
Adhesion value in N cm^{-1} after ageing	0.84	0.63	0.26	0.62
Wet strength $\text{N} \cdot \text{cm}^{-1}$	1.28	0.62	0.08	0.54

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CLAIMS

1. A sprayable hotmelt adhesive with a viscosity of 500 to 4,000 mPas at 150°C, characterized by the following composition:
- 5 A) 30 to 70% by weight of at least one poly- α -olefin or poly- α -olefin mixture with a softening point (ring-and-ball method) of 70 to 130°C and a melt viscosity at 190°C of 1,000 to 20,000 mPas,
B) 5 to 30% by weight of at least one oil,
C) 20 to 60% by weight of at least one hydrocarbon resin with a softening
10 range of 70 to 140°C and
D) optionally additives.
2. A hotmelt adhesive as claimed in claim 1, characterized by a viscosity of 700 to 1,900 mPas at 150°C, as measured in accordance with
15 ASTM D 3236-88.
3. A hotmelt adhesive as claimed in claim 1, characterized in that the poly- α -olefin or the poly- α -olefin mixture is substantially amorphous and the poly- α -olefin has the following monomer composition:
- 20 - 3 to 75% by weight of an α -olefin containing 4 to 10 carbon atoms,
- 25 to 95% by weight of propene and
- 0 to 20% by weight of ethene.
4. A hotmelt adhesive as claimed in claim 1, characterized in that the poly- α -olefin or the poly- α -olefin mixture has a melt viscosity at 190°C of 2,000 to 15,000 mPas.
- 25 5. A hotmelt adhesive as claimed in claim 1, characterized in that the poly- α -olefin has a density of <0.90 g/cm³, a needle penetration of 8 to 4.0 mm, a molecular weight as determined by gel permeation chromatography of at most 100,000 (weight average) or at least 4,000 (number average), the difference between the weight average and the number average
30 molecular weight being no more than six times the number average.

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6. A hotmelt adhesive as claimed in claim 1, characterized in that the poly- α -olefin mixture contains at least one poly- α -olefin with a melt viscosity of 40,000 to 60,000 and at least one poly- α -olefin with a melt viscosity of 3,000 to 10,000 Pas at 190°C.
- 5 7. A hotmelt adhesive as claimed in claim 1, characterized in that the paraffinic oil is a medicinal white oil.
8. A hotmelt adhesive as claimed in claim 1, characterized in that the hydrocarbon resin is a hydrocarbon resin containing 5 to 9 carbon atoms.
9. A hotmelt adhesive as claimed in claim 1, characterized in that the additive is at least one substance of the following group: heat and light stabilizer, optical brightener, antistatic agent, lubricant and antiblocking agent, nucleating agent, dye, pigment or flame retardant.
- 10 10. A hotmelt adhesive as claimed in claim 1, characterized in that components B and C together make up at least 30% by weight, preferably at least 35% by weight and more preferably at least 45% by weight of the sum of components A+B+C.
- 15 11. A hotmelt adhesive as claimed in claim 1, characterized in that components A, B and C are selected so that the viscosity at 100°C is in the range from 5 to 15 Pas \pm 15, more particularly \pm 10%, as a function of the shear rate of 2 to 250 [sec⁻¹].
- 20 12. A process for the production of the hotmelt adhesive claimed in at least one of claims 1 to 11, characterized in that the raw materials are mixed in an inert gas atmosphere and/or in a vacuum at temperatures of 150 to 200°C.
- 25 13. The use of the hotmelt adhesive claimed in at least one of claims 1 to 11 for structural bonding in sanitary products, more especially for bonding diapers, panty liners and sanitary napkins.
14. The use of the hotmelt adhesive claimed in at least one of claims 1 to 11 for bonding films, more particularly of polyolefins, and nonwovens, more particularly of polypropylene, the application temperature being
- 30

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between 120 and 180°C and preferably between 140 and 160°C, the coating weight being between 2 and 10 and preferably between 3 and 4 g/m² and the application rate preferably being between 50 and 400 m/min.

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Abstract of the Disclosure

The present invention relates to a sprayable hotmelt adhesive composition, a process for preparing the same, and a method of using the same. The hot melt adhesive composition contains 30 weight percent to 70 weight percent of one or more poly- α -olefins; 5 weight percent to 30 weight percent of at least one oil; and 20 weight percent to 60 weight percent of at least one hydrocarbon resin having a softening range of 70°C to 140°C. The poly- α -olefin or the mixture of poly- α -olefins has a softening point of 70°C to 130°C and a melt viscosity at 190°C of 1,000 mPas to 20,000 mPas. The hotmelt adhesive has a viscosity at 150°C of 500 mPas to 4,000 mPas.

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DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION

☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial Filing

Attorney Docket Number **H 3381 PCT/US**

First Named Inventor **Kolowrot, Dirk**

COMPLETE IF KNOWN

Application Number **09/787,248**

Filing Date

Group Art Unit

Examiner Name

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

SPRAYABLE HOT-MELT ADHESIVE

(Title of the Invention)

the specification of which

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY) **09/14/1999** as United States Application Number or PCT International

Application Number **PCT/EP99/06799** and was amended on (MM/DD/YYYY) (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code § 119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached?	
			YES	NO	
198 43 141.4	Germany	09/21/1998	<input type="checkbox"/>	<input type="checkbox"/>	<input checked="" type="checkbox"/>
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U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP99/06799	09/14/1999	

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				ADDITIONAL INVENTOR(S) Supplemental Sheet			
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Name of Additional Joint Inventor, if any:				<input type="checkbox"/> A petition has been filed for this unsigned inventor			
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